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Model and Accuracy

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ATMOSPHERIC MODELING: MODEL AND ACCURACY

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The development of models to assess the emission control requirements of primary precursor pollutants in the production of photochemical oxidants has been underway for approximately 20 years. Over this period there has been a considerable increase in our understanding of the basic scientific processes associated with the production of photochemical oxidants in the atmosphere and a concomitant advance in the analytical sophistication by which these processes are incorporated to derive quantitative precursor relationships. The establishment of air quality standards implies the use of such quantitative relationships to describe the link between emissions and air quality in order to consider an effective and cost efficient control program. This paper maps out the progress that has been made in this endeavor, discussing the successes and shortcomings of the present generation of modeling tools. Model performance evaluations, accuracy, uncertainty and sensitivity analyses are also discussed.

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16. ABSTRACT

The development of models to assess the emission control requirements of primary precursor pollutants in the production of photochemical oxidants has been underway for approximately 20 years. Over this period there has been a considerable increase in our understanding of the basic scientific processes associated with the production of photochemical oxidants in the atmosphere and a concomitant advance in the analytical sophistication by which these processes are incorporated to derive quantitative precursor relationships. The establishment of air quality standards implies the use of such quantitative relationships to describe the link between emissions and air quality in order to consider an effective and cost efficient control program. This paper maps out the progress that has been made in this endeavor, discussing the successes and shortcomings of the present generation of modeling tools. Model performance evaluations, accuracy, uncertainty and sensitivity analyses are also discussed.

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INTRODUCTION

The first oxidant control strategies recommended by the U.S. Environmental Protection Agency were based on ambient pollutant concentration data taken in several U.S. cities. These data were analyzed to relate the daily one hour averaged maximum oxidant concentration to the average nonmethane hydrocarbon (NMHC) concentration observed in the 6 to 9 a.m. time period.[1,2] This so called upper-limit curve, which inherently assumes that hydrocarbons are the unique dependent variable in oxidant control, became the basis for calculating the control requirement necessary to meet the National Ambient Air Quality Standard for oxidants. The resulting oxidant - NMHC relationship, referred to as Appendix J., was determined to be scientifically incomplete based on evolving knowledge of atmospheric photochemical oxidation processes[3] and the meteorology of urban boundary layers.[4]

With improved knowledge has come the development of a variety of improved scientific approaches for quantitatively treating the relationship between precursor emissions and ozone formation. Among others, these include, in order of increasing meteorological sophistication: 1) the Empirical Kinetics Modeling Approach;[5] 2) the Photochemical Box Model;[6,7] 3) the Lagrangian Photochemical Model;[8] and 4) the Urban Airshed Model.[9] All these advanced techniques incorporate state of the art photochemical mechanisms. Some differences among the techniques are introduced as a result of the assumptions applied in treating the complex chemical composition of polluted atmospheres, but more significant are the differences that appear in the level of treatment of the meteorological processes in the approaches.

THE MODELS

Models, in the context of this paper, are defined as any set of mathematical equations that relate pollutant emissions to ambient air quality through empirical relationships and/or theoretical descriptions of the chemical and physical processes occurring in the atmosphere. As such, the models consider the following processes in varying degrees of sophistication: 1) chemical and physical transformations; transport and diffusion; and dry and wet removal of atmospheric pollutants.

The Empirical Kinetics Modeling Approach (EKMA), unlike the other models, does not consider meteorological processes explicitly, but does consider what might be viewed as a generic worst case meteorological scenario. In addition EKMA does not make absolute predictions of ambient oxidant levels, but predicts instead relative changes in ozone levels as a function of precursor emission reductions. The method uses historical ambient observations of non-methane hydrocarbons and oxides of nitrogen in the early morning and the resulting maximum ozone level for a worst case episodic day in the urban area of interest. As a result, the technique is not amenable to performance evaluations involving comparative analysis of observations and predictions.

The Photochemical Box Model (PBM) is a variable volume, well mixed reaction cell which simulates air pollutant concentrations in urban atmospheres for spatial averages over the volume of the cell. Meteorological considerations include variation of solar insolation as a function of time, time of the year, and cloud cover; transport of pollutants into and out of the modeling domain; and a temporally resolved vertical mixed layer.

The Lagrangian Photochemical Model (LPM) is a multi-level moving reaction cell which simulates air pollutant concentrations in urban atmospheres over a spatial pathway defined by the air parcel's trajectory driven by the horizontal components of the wind. In addition to the features of temporally varying solar insolation and mixing height available in the PBM, the LPM provides for vertical resolution of pollutant concentrations within the air parcel. The model has limitations in resolving horizontal concentration fields due to the impracticality of running the thousands of trajectory calculations necessary to generate such fields.

The Urban Airshed Model is a multi-level, multi-cell grid approach which simulates air pollutant concentrations over the entire urban domain at a fixed specified horizontal grid resolution (usually 4- 10 km). This is the most sophisticated of the approaches, encompassing all the features of the previously mentioned models with the added capability of providing three dimensional pollutant concentration fields.

The chemical characteristics of the pollutant species of interest, to a large extent, govern the relative importance of the processes mentioned above. For example, in the case of modeling ozone episodes in urban environments, the time scale of interest is of the order of a diurnal cycle (~12 hrs). During periods of high solar insolation and limited atmospheric ventilation of the urban area , the production of ozone is rapid. The fact that removal of pollutant gases by dry deposition is a relatively slow process; its impact on modeling ozone on the urban scale is minor. In the case of wet removal, the process is not of interest, since precipitating days lack the solar intensity to drive the photochemical smog formation process. Neither of these removal processes can be considered unimportant when considering oxidant and its precursors on the regional scale, that is, transport scales of the order of 1000 km and chemical and physical process time scales of the order of several days. It should also be noted that regional contributions (lateral and top boundary inflow) of ozone to urban episodes are significant under many atmospheric conditions and must be considered in the analyses performed.

MODEL EVALUATION

It is generally recognized that the most significant single step in building confidence in a model's validity and predictive capability is through statistical performance tests based on comparisons of model predictions with atmospheric observations of pollutant concentrations. Performance measures for models have been suggested by the American Meteorological Society[11] and others[10,12] and several evaluation studies[13-15] have been completed for photochemical oxidant models. A major deterent in such evaluations has been the availability of adequate urban data bases. This situation is slowly improving as monitoring networks and emission inventories in urban areas evolve, and as technical expertise in applying these sophisticated models develops operationally in State air quality management organizations.

One of the most ambitious model evaluation programs to date was the St. Louis Regional Air Pollution Study (RAPS) sponsored by the U.S. Environmental Protection Agency. In this extensive monitoring and research program three modeling approaches were tested with a twenty day sample size taken from a two year monitoring record collected in St. Louis, Missouri. A summary of the results is presented in Table I.

The evaluated models had standard deviations of differences between observed ozone maxima and predicted concentration ranging from 39 to 58 ppb. The observed maxima for the twenty day evaluation set ranged from 160 to 260 ppb. Overall model predictions were within plus or minus 30% of observations and if the temporal and spatial specificity requirements for the model predictions were relaxed, the performance was further improved to plus or minus 10%. Other evaluations considering urban airshed type models in Los Angeles, CA[14] and Denver, CO[15], but for a smaller sample set, showed performance statistics consistent with those reported in the St. Louis evaluations.

It is important to note that though the performance evaluations build confidence in the model's fidelity in treating the important physical and chemical processes relevant to the formation of photochemical oxidants in polluted atmospheres, the evaluations are not a direct test of the model's predictive capability in an operative sense. That is, the model's performance in predicting a change in maximum ozone concentration as a function of a change in precusor emissions has not been tested. This more definitive model performance evaluation has been impeded mainly due to a lack of adequate data bases (data records of 5 or more years for ozone and its precursors and the corresponding emission inventories) for running the models.

MODEL SENSITIVITY AND UNCERTAINTY ANALYSIS

In addition to the progress being made in model evaluation, significant advances in the development and application of sensitivity analysis techniques to air quality simulation models have been achieved. The terms sensitivity and uncertainty analysis are sometimes used ambiguously. They are defined explicity in the context of this discussion as follows.

Sensitivity analysis, as used here, refers to determining the nature and extent of the change in model output that results from a variation in model input; where model input is used in its broadest sense. That is, we consider not just the observational and empirical data used to drive a given set of processes defined by a particular modeling system, but also the parameterizations and assumptions used in the model to define the processes themselves.

Uncertainty analysis refers to determining the limits of variation in model prediction as a result of the approximate nature of the modeling system and limits in our ability to precisely describe the chemical and meteorological state of the atmosphere for input into the model.

For example, in the sensitivity analyses of urban photochemical models[4,16], it has been demonstrated that model inputs of initial and boundary concentration conditions, mixing height variations, and photolysis intensity have significant impact on model predictions. Through sensitivity analyses of photochemical mechanisms[17,18], it is possible to identify critical rate constants and fragmentation processes which affect the yields of ozone. This then provides an estimate (mostly likely a lower limit) of the associated uncertainties in predicting precursor control requirements using presently available photochemical models. It is important to note that sensitivity analysis pertains to the sensitivity of the model. It is theoretically based, and does not necessarily reflect the actual response of the perturbed atmosphere.

To empirically perform a model uncertainty analysis, requires a rather extensive observational data set encompassing a broad range of meteorological and chemical conditions. Such data sets will allow extensive comparisons of model predictions with observations to generate probability distribution functions of the differences between modeled and observed values. The difficulty with such comparisons is that the sources of uncertainty (for example, disparities in spatial representativeness between model predictions and observations, observational errors, or uncertainties propagated in the model from inputs) are not resolvable. It may be possible through selective partitioning of the data sets to reduce some of the existing degrees of freedom and augment the resolution of the uncertainty analysis.

Theoretical sensitivity analysis techniques can also be used to incorporate the uncertainty distribution of the input parameters to generate the probability distribution function of the model predictands, recognizing the limitations previously mentioned (see for example Morgan et al.[19]). The difficulty with implementing these techniques is the result of limited data for use in developing objective probability density functions for key model parameters and establishing their joint probablity distributions. A complete model uncertainty analysis has not been performed to date, primarily due to a lack of adequate data sets.

CONCLUSIONS

Progress continues to be made in the physical and chemical characterization of the complex processes associated with the production of oxidants/O_3 in polluted atmospheres. Our goal is to provide tools based on sound, credible scientific principles which relate primary precursor emissions to oxidants/O_3 products. As a result of continuing research and development efforts, significant advances in our understanding of the complex processes operating in photochemical smog have been made. These include improved mechanisms for treating the photochemical oxidant cycle, better understanding and treatment of urban boundary layer processes, initiation of systematic model performance evaluation studies, advances in sensitivity and uncertainty analysis techniques and their application and consideration of oxidants/O_3 on regional scales (~ 1000 km). In addition, the process of determining the nature and extent of the uncertainty associated with model prediction has begun. The recognition of model uncertainty and its estimation, requires that the scientific community make a special effort to better articulate the limitations in model predictability and to provide a better understanding of how these limits can be incorporated effectively in the decision making process.

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Table I.
Summary of St. Louis RAPS Model Evaluation Study

	Statistics on max hourly [O3] for highest 20 days in 1975 -1976		
	$\overline{\Delta C}$	s.d.($\overline{\Delta C}$)	$\lvert\overline{\Delta C}\rvert$
Photochemical Box Model	-0.012	0.039	0.029
Lagrangian Photochemical Model	0.023	0.058	0.052
Urban Airshed Model	-0.062	0.035	0.062

*ΔC = Obs. - Pred. (ppm)